

Depth dependent spin dynamics of canonical spin glass films: A low-energy muon spin rotation study

E. Morenzoni,^{1,*} H. Luetkens,^{1,2} T. Prokscha,¹ A. Suter,¹ S. Vongtragool,^{1,3}
F. Galli,³ M.B.S. Hesselberth,³ N. Garifanov,⁴ and R. Khasanov^{1,5}

¹*Paul Scherrer Institut, Labor für Myon-Spin Spektroskopie, CH-5232 Villigen PSI, Switzerland*

²*Institut für Physik der Kondensierten Materie, TU Braunschweig, D-38106 Braunschweig*

³*Kamerlingh Onnes Laboratory, Leiden University,*

P.O.B. 9504, 2300 RA Leiden, The Netherlands

⁴*Kazan Physicotechnical Institute, RAS, Kazan 420029, Russia*

⁵*Physik Institut der Universität Zürich, CH-8057 Zürich, Switzerland*

(Dated: February 20, 2008)

We have performed depth dependent muon spin rotation/relaxation studies of the dynamics of single layer films of *AuFe* and *CuMn* spin glasses as a function of thickness and of its behavior as a function of distance from the vacuum interface (5-70 nm). A significant reduction in the muon spin relaxation rate as a function of temperature with respect to the bulk material is observed when the muons are stopped near (5-10 nm) the surface of the sample. A similar reduction is observed for the whole sample if the thickness is reduced to e.g. 20 nm and less. This reflects an increased impurity spin dynamics (incomplete freezing) close to the surface although the freezing temperature is only modestly affected by the dimensional reduction.

PACS numbers: 75.50.Lk, 75.70.Ak, 76.75.+i, 75.30.Hx

Spin glasses are founded on the randomness and frustration of the exchange interaction between diluted spins embedded in a non-magnetic environment. Their dynamics as a prototype of the dynamics of glassy and disordered systems with complex phase space has remained a topical issue for many years [1, 2, 3]. Although significant progress has been made in understanding basic properties of the spin relaxation above and below the freezing temperature T_f , which appears as a cusp like peak in the zero field cooled (ZFC) susceptibility [4], the exact nature of the spin glass ground state, of its ordering, and of the low lying excitations are still controversial and remain an unsettled theoretical problem [5].

The random freezing of the magnetic moments, which involves strong cooperative effects, is expected to exhibit finite size effects and many theoretical investigations have focussed on the question of the lower critical dimension [6]. From the experimental point of view it is therefore important to investigate whether the dynamics of the spin glass shows a different pattern if one sample dimension (e.g. the thickness) is reduced or whether it is homogenous throughout the sample. The experimental difficulty to obtain an observable magnetization signal for layer thicknesses in the nm range has led to many investigations of multilayered samples consisting of spin glass layers separated by decoupling layers. Measurements extending down to sub nanometer layers of *CuMn* and *AgMn* found a decrease of T_f below a thickness of 100 nm with a significant drop only below a few nanometers and still a finite T_f at one monolayer, indicating that even at this thickness a canonical spin glass displays the characteristic macroscopic signatures [7]. ZFC magnetization and AC susceptibility measurements have shown a

pronounced relative shift of the frequency dependent T_f upon decreasing the thickness from bulk to about 2 nm and were interpreted in the frame of crossover from 3D to 2D dynamics [8].

There are only a few techniques applicable to single layer spin glasses where possible complications of multilayer samples such as interlayer diffusion, inhomogeneity and role of decoupling layers can be avoided. Resistance noise measurements have been used as an alternative to magnetic measurements [9]. Measurements of the anomalous Hall effect detected a large reduction of the out of plane magnetization in *AuFe* films (30nm) with respect to the bulk which was explained as originating from the surface anisotropy of the impurity spin magnetization introduced by the presence of the vacuum interface [10]. A new technique able to provide local information about static fields and dynamic fluctuations of magnetic moments is now offered by the availability of a polarized low-energy muon beam at the Paul Scherrer Institute, which allows depth dependent muon spin rotation/relaxation (μ SR) investigations in thin single layer spin glasses [11].

In this letter we report on studies of single layers (10, 20, 50 and 220 nm) of the canonical spin glasses *AuFe* 2.2 and 3 at.%, *CuMn* 2 at.% and of a double layer *AuFe*(31nm)3 at.% on *Au*(160nm). The polarized muons with their spin precessing and depolarizing in the neighborhood of the magnetic moments act as microscopic local probes of statics and dynamics of these moments. They are very well suited for the study of spin glasses, because of their high sensitivity in the time window of magnetic fluctuations of 10^{-4} - 10^{-10} sec [13]. The time evolution of the polarization of the muon ensemble $P(t)$

is observed via detection of the emitted decay positron intensity as a function of time after implantation. By varying the energy of the low-energy muons we can follow the temperature dependence of the muon spin relaxation as a function of layer thickness and of depth below the surface. We find pronounced thickness and depth dependent effects that indicate an "incomplete" freezing of the magnetic moments in a ~ 10 nm region below the surface interfacing the vacuum.

Muon-spin relaxation has provided novel insight about the nature of magnetic freezing in bulk systems ([12, 13] and references therein). The magnetic field at the muon site is mainly caused by the dipolar fields of the magnetic ions. First experiments in an external magnetic field transverse (TF) to the initial muon spin direction showed a rapid relaxation increase when T_f is approached from above [14]. At temperatures well above T_f the electronic moments of the magnetic impurities are rapidly fluctuating so that the resulting depolarization, given by the field averaged over the muon life-time (2.2 μ sec), is small. Cooling the spin glass down to its freezing temperature slows down the fluctuations, thereby bringing the fluctuation rate into the time window of the muon and increasing the observable depolarization. Below T_f , the electronic moments become static on the scale of the muon life-time, causing the muons to precess in the static dipolar fields of the magnetic ions. In a translational invariant ferromagnet or antiferromagnet a single precession frequency may be observed. However, in a spin glass the dipolar fields are random in magnitude and direction when averaged over the sample as in a μ SR experiment and only a fast depolarization is observable. Zero (ZF) and longitudinal field (LF) measurements clearly demonstrated the coexistence of static and dynamic random local fields and the gradual build up of a static moment below T_f reflecting homogenous freezing [13]. Further studies relating $P(t)$ to the local spin autocorrelation function $\langle \vec{S}(t) \cdot \vec{S}(0) \rangle = q(t)$ and its scaling behavior as a function of the applied field showed that above and close to T_f the correlations of the impurity moments are strongly non-exponential [15].

Our samples (size 25 x 25 mm²) have been prepared by co-sputtering high purity metals onto a Si/SiO₂ substrate. Before the argon sputter gas was admitted the equipment was pumped down to UHV conditions (10^{-8} mbar) to avoid oxidation of the films during fabrication. The films were analyzed using Rutherford backscattering (RBS) and electron microprobe analysis. RBS confirmed a homogenous depth distribution of the magnetic impurities. Simultaneously, narrow stripes for resistivity and Hall-effect measurements for T_f determination were sputtered. Thicker films prepared for magnetic susceptibility measurements confirmed the freezing temperatures reported in the literature for the concentration chosen. Within the error the resistivity of the films (about twice as large as in the bulk material) does

not depend on the thickness indicating the equivalence of the disorder in all films. Muons with energies between 1 and 22.5 keV were implanted in the samples. The range profiles were calculated using the Monte Carlo program TRIM.SP [16, 17]. Over the investigated temperature range the muons do not diffuse and randomly occupy octahedral interstitial sites in the fcc-lattice. We performed the experiments in ZF and in 10 mT (TF). A transverse field influences properties of the spin glass such as the sharpness of the transition, but leaves unaltered the essential features reflecting the depth and thickness dependent dynamical behavior of the films. Also previous μ SR and NMR investigations are consistent with an essentially field independent spectral function $J(\omega)$ of the fluctuating fields [18]. The observed $P(t)$ was analyzed by fitting with a stretched exponential relaxation function, $\exp[-(\lambda t)^\beta]$ (multiplied by a precession factor $\cos(\omega_\mu t)$ for the TF measurements). This function has been found appropriate in cases where a complex non-exponential fluctuation pattern is expected [19]. Additionally we analyzed the ZF measurements with a muon spin relaxation function, which is able to describe the coexistence below T_f of static and dynamic random fields produced by the impurity moments [13]:

$$P(t) = P(0) \left[\frac{1}{3} e^{-\sqrt{\lambda_d} t} + \frac{2}{3} \left(1 - \frac{a_s^2 t^2}{\sqrt{\lambda_d t + a_s^2 t^2}} \right) e^{-\sqrt{\lambda_d t + a_s^2 t^2}} \right]. \quad (1)$$

Here $a_s = a_0 \sqrt{Q}$ is the static field width probed by the muons, a_0 is the width corresponding to full static moments at $T=0$ and Q is the Edwards-Anderson order parameter describing the non vanishing part of the impurity spin autocorrelation function for infinite time [2]. The dynamical relaxation introduced by the fluctuations is described by the parameter λ_d . The results for the muon spin relaxation rate λ and exponent β as a function of temperature are shown in Fig. 1 for single layers of AuFe of various thicknesses (the CuMn system gives similar results). The effect of the reduced sample dimension is evident. For each thickness, $\lambda(T)$ increases steadily on approaching T_f from above, reflecting the slowing down of the moments. In the 50 nm sample the increase is similar to the one observed for bulk samples. However, already in the 20 nm layer a reduction of $\lambda(T)$ is visible. This reduction is much more pronounced in the 10 nm layer so that well below T_f $\lambda \lesssim 1 \mu s^{-1}$ less than 10% of the 50 nm value; this factor cannot be accounted for by a pure geometric effect on the static width of the spin glass surface. The exponent β reaches 1 (corresponding to a single correlation time) only well above T_f and decreases towards $\frac{1}{3}$ at low temperatures, a behavior qualitatively observed in bulk samples. The large reduction of the relaxation rate in the thin films points to the persistence of spin fluctuations at low temperatures. This can be due to a finite size or to a surface effect. In the latter case the

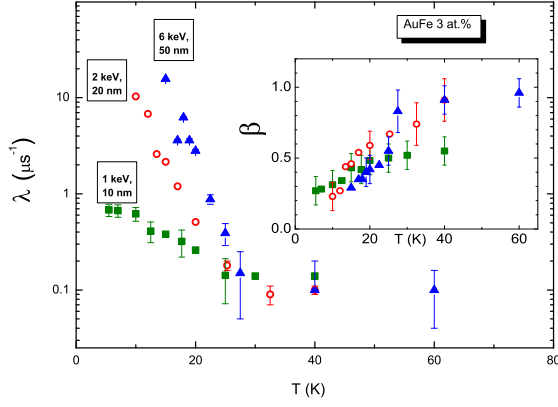


FIG. 1: Muon depolarization rate (λ) and stretch exponent (β , see inset) versus temperature for *AuFe* thin films of 10, 20 and 50 nm, 10 mT TF data. Muons of 1, 2, 6 keV respectively, are stopped in the center of the layer.

effect should emerge also in a thicker sample. We have therefore performed depth dependent investigations on *AuFe* 3 at.% 50 and 220 nm films by varying the implantation energy between 1 keV (mean depth \bar{d} =6 nm) and 22.5 keV (\bar{d} =68 nm).

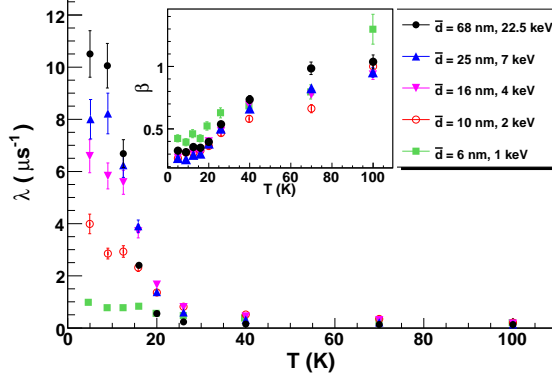


FIG. 2: Muon depolarization rate (λ) and stretch exponent (β , see inset) as a function of temperature (T) at different (mean) depths in *AuFe* 3 at.%. Thickness: 220 nm, ZF data.

Results are plotted in Fig. 2. Clearly, λ is depth dependent. A strong reduction is found at 2 keV and below corresponding to stopping profiles centered in the top 10 nm of the spin glass. Again similar behavior is observed in *CuMn*. The results imply that the reduction of the muon spin relaxation in the 20 nm and the strong suppression in the 10 nm sample are a manifestation of the dominant contribution of the surface layer at these thicknesses. In principle a smaller λ in the TF and ZF measurements can be due to a pure reduction of the static width below T_f or to enhanced dynamics and consequent motional narrowing or a combination of both. At the moment our apparatus does not allow routine LF investigations in a large range of fields to follow the repolarizing effect typi-

cal of the pure static case. However, LF measurements in the 10 nm *CuMn* sample in fields up to ~ 13 mT at 4 K do not show any change of $P(t)$. The fact that this LF does not decrease relaxation rates of $\lesssim 1\mu s^{-1}$ (corresponding to $\lesssim 1$ mT static field width) indicates dynamical effects as a cause of the observed behavior. This is also confirmed by an heuristic interpretation of the β and λ parameters. Formally, a stretched exponential $P(t)$ can be expressed as a distribution of exponential muon relaxation rates λ_i each reflecting the contribution of local spins relaxing with a time $\tau_i \propto \lambda_i^{-1}$ [19]. The distribution function of λ_i reflects the weighting distribution $g(\tau_i)$ of the corresponding exponential autocorrelation functions with β and λ determining the shape of $g(\tau_i)$. Reducing β below 1 widens the distribution. In addition lowering λ shifts the weight from long correlation times, which appear as static component to the μ^+ , to very short ones. The depth dependence of λ for $T < T_f$ reflects therefore a wide distribution of spin relaxation times with persistence of a substantial dynamic component close to the surface.

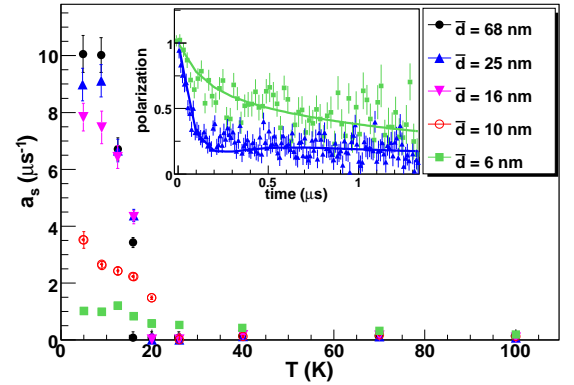


FIG. 3: Static amplitude of random fields a_s in the *AuFe* film (thickness 220 nm) at different depths. In the inset polarization spectra (normalized to the asymmetry obtained in pure Au) at 5K and 2 different depths are shown. The curves are fit to Eq. 1.

The analysis of the ZF data with Eq. 1 allows to separate static from dynamic contributions. A pronounced depth dependence is observed in the magnitude of the static field component, a_s (Fig. 3), which attains finite values below $\sim T_f$ to saturate on lowering the temperature. Overall the curve shows that this order parameter is gradually reduced in the ~ 10 nm layer below the surface. This is clearly visible in the shape of $P(t)$ taken at 5 K and two different depths of the 220 nm film (Fig. 3 inset). Whereas well below the surface $P(t)$ exhibits a rapid decrease followed by a tail characteristic of the presence of (pronounced) static field components, close to the surface weak relaxation dominates the spectrum. The dynamic component λ_d peaks around $\sim T_f$ in agreement with bulk results [13]. The reduction of a_s below T_f and the behav-

ior of λ_d means that rapid large fluctuations with typical correlations times $\tau \ll \frac{1}{a_s} = 10^{-6}s$ around a preferred direction do not freeze even at the lowest temperature. It should be remarked that the increased dynamics is not accompanied by a concomitant sizeable reduction of T_f . This is visible in Fig. 3 which evidences the strong variation of a_s with depth and a within errors practically unchanged T_f as determined from $a_s(T_f) = 0$. Also in spin glasses of *AuFe*, *AgMn* and *CuMn* at higher magnetic concentration the reduction of the thickness to 10 nm was found to reduce the freezing temperature by at most 30% [7, 9]. Seynaeve et al. [10] observed a strong decrease of the magnitude of the anomalous Hall effect as the thickness of the spin glass gets smaller and a disappearance of the effect when their *AuFe* film was sandwiched between layers of pure Au. Both observations were interpreted as the result of in-plane surface anisotropy of the static Fe-moments caused by spin-orbit scattering of electrons by the non-magnetic host atoms. Our μSR measurements of a double layer *AuFe*(31 nm)/*Au*(160 nm) also exhibit a reduction of the low-temperature relaxation near the *AuFe* / vacuum interface, but not near the *AuFe* / *Au* interface. However, surface anisotropy as the only cause of the observed thickness and depth behavior can be ruled out since in the dilute case randomly oriented or aligned dipoles both produce Lorentzian field distributions with the same width [20] and would leave the μSR spectra and parameters unmodified. We confirmed this by numerical simulations. On the other hand our results, showing the presence of a fluctuation spectrum with large amplitude oscillation below T_f , can explain the Hall effect measurements of [10].

In conclusion we have used the sensitivity and the depth profiling capacity of LE- μSR to study the inhomogeneous magnetization and dynamic profile in dilute magnetic alloys. We find that on a length scale of ~ 10 nm the surface of canonical spin glass systems exhibits a considerably enhanced fluctuating behavior in comparison with the bulk, whereas the apparent freezing temperature does not change dramatically. This reduction of the order parameter modifies the magnetic state of films of comparable thickness. To our knowledge no theoretical model is able to predict this inhomogeneous behavior or to identify the underlying physical mechanism. However, recent calculations of the low energy excitations of the Edwards-Anderson model of Ising spin glasses find that a dimensional reduction of 10-30% of T_f is accompanied by a disproportionately larger reduction of the stiffness exponent until the lower critical dimension $d=\frac{5}{2}$ is reached [6]. Since this exponent governs the typical energy scale for excitation, lowering it reduces the resistance against the formation of low energy excitations, a behavior in qualitative accordance with our observation.

It has been predicted that in an inhomogeneous situation such as the one represented by the presence of a vac-

uum interface the RKKY interaction, which determines the interaction between the impurity spins, may be modified [21] but its effect on the spin dynamics or freezing behavior remains to be quantified. Recent experiments [22] have shown that the surface of ferromagnetic layers is weakly exchange coupled to the bulk, resulting in faster dynamics of the surface magnetization but contrary to the spin glass in the ferromagnetic case only a surface layer of sub nm thickness is concerned. On the other hand, the length scale of enhanced fluctuations observed here may explain why in previous multilayer experiments decoupling layers of several tens of nm were necessary to observe dimensional effects [23]. We hope that the results presented here will stimulate theoretical calculations of the spin dynamics in the situation where translational invariance is not satisfied.

This work was performed at the Swiss Muon Source S μ S, Paul Scherrer Institute, Villigen, Switzerland and financially supported by FOM, The Netherlands. We thank G. Nieuwenhuys for the important contributions in the early stage of this work.

* E-Mail:Elvezio.Morenzoni@psi.ch

- [1] K. Binder, A.P. Young, Rev. Mod. Phys. **58**, 801 (1986).
- [2] K.H. Fisher, J.A. Hertz, *Spin Glasses*, Cambridge University Press, Cambridge (1991).
- [3] *Spin Glasses and Random Fields*, edited by A.P. Young, World Scientific, Singapore (1998).
- [4] J.A. Mydosh, *Spin Glasses: An Experimental Introduction*, Taylor and Francis, London (1993).
- [5] M. Palassini, A. P. Young, Phys. Rev. Lett. **85**, 3017 (2000).
- [6] S. Boettcher, Phys. Rev. Lett. **95**, 197205 (2005).
- [7] L. Hoines *et al.*, Phys. Rev. Lett. **66**, 1224 (1991).
- [8] L. Sandlund *et al.*, Phys. Rev. **B40**, 869 (1989).
- [9] P.W. Fenimore, M.B. Weissman, J. Appl. Phys. **85**, 8317 (1999).
- [10] E. Seynaeve *et al.*, Phys. Rev. Lett. **85**, 2593 (2000).
- [11] E. Morenzoni *et al.*, J. Phys.: Cond. Matt. **16**, S4583-S4601 (2004).
- [12] R.H. Heffner *et al.*, J. Appl. Phys. **53**, 2174 (1982).
- [13] Y. Uemura *et al.*, Phys. Rev. **B31**, 546 (1985).
- [14] D.E. Murnick *et al.*, Phys. Rev. Lett. **36**, 100 (1976).
- [15] A. Keren *et al.*, Phys. Rev. **B64**, 054403 (2001).
- [16] W. Eckstein, *Computer Simulations of Ion-Solid Interactions*, (Springer Verlag Berlin, Heidelberg and New York, 1991).
- [17] E. Morenzoni *et al.*, Nucl. Instr. and Methods **B192**, 254 (2002).
- [18] D.E. MacLaughlin *et al.*, Phys. Rev. Lett. **51**, 927 (1983).
- [19] I.A. Campbell *et al.*, Phys. Rev. Lett. **72**, 1291 (1994).
- [20] D.V. Berkov, Phys. Rev. **B53**, 731 (1996).
- [21] J.S. Helman, W. Baltensperger, Phys. Rev. **B50**, 12682 (1994).
- [22] F. Sirotti *et al.*, Phys. Rev. **B61**, R9221 (2000).
- [23] P. Granberg *et al.*, Phys. Rev. **B44**, 4410 (1991).